Zn-doping dependence of the wipeout region around Zn in $YBa_2(Cu_{1-x}Zn_x)_4O_8$

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We estimated the spread of the magnetically enhanced regions (wipeout regions) around Zn ions in high-T_c superconductors $YBa_2(Cu_{1-x}Zn_x)_4O_8$ (x=0, 0.005, 0.01, and 0.022) at T=4.2 K, via the planar ^{63}Cu nuclear spin-lattice relaxation study with Cu nuclear quadrupole resonance (NQR) spin-echo technique. From the analysis of nonexponential planar Cu nuclear spin-lattice relaxation curves, we found that the wipeout region per a Zn ion shrinks with Zn doping in the superconducting state. The shrinkage is associated with suppression of the host antiferromagnetic spin correlation, as a result from a dilution effect on the magnetic CuO_2 network.

76.60.-k, 74.25.Nf, 74.72.-h

I. INTRODUCTION

Nonexponential relaxation is frequently observed in a wide class of disordered or inhomogeneous materials. For nonmagnetic impurity Zn-doped high-T_c superconductors $YBa_2Cu_3O_{7-\delta}$, the nonexponential planar Cu nuclear spin-lattice relaxation curves are observed [1]. A lot of effort has been made to understand how the nonexponential recovery curves are described and what is the physics behind it. Recently, it has been shown that the magnetic impurity-induced NMR/NQR relaxation theory well reproduces the observed recovery curves for impurity-doped high-T_c superconductors [2–6], which includes the relaxation process due to a host Cu spin-fluctuation, that due to an impurity-induced spin-fluctuation, and a wipeout effect [7,8]. The wipeout effect is defined as a loss of NMR/NQR signal more than what would be expected from simple dilution effect due to substitution of the foreign atoms. The foreign atom is assumed to cause a field gradient sufficiently large to shift the resonance frequency of the surrounding nuclei outside the observable range, or to cause a local field fluctuation sufficiently large to diminish the NMR/NQR signal in the observable time domain. The wipeout effect on Cu NQR spectra has been observed in the normal state of $YBa_2(Cu_{1-x}Zn_x)_3O_{7-\delta}$ [9].

In this paper, we estimated the wipeout number N_c in the superconducting state as a function of Zn content, which could not be estimated from measurements of the Cu NQR spectra, via the planar ^{63}Cu nuclear spin-lattice relaxation study for $YBa_2(Cu_{1-x}Zn_x)_4O_8$ (Y124). In the light of the impurity-induced NQR relaxation theory with the wipeout effect [7,8], we found shrinkage of the wipeout region around each Zn ion with Zn doping.

II. EXPERIMENTAL

Powder samples of the Zn-doped Y124 (x=0.005, 0.010 and 0.022; T_c=68, 56 and 15 K) for the Cu NQR experiments are the previously studied ones in Refs. 3-6. Zero-field Cu NQR measurements were carried out with a coherent-type pulsed spectrometer. Nuclear spin-lattice relaxation was measured by an inversion recovery spin-echo technique, where the ^{63}Cu(2) nuclear spin-echo intensity $M(t)$ was recorded as a function of the time t after an inversion pulse. For comparison, the same experiments have been made for the optimally carrier-doped $YBa_2(Cu_{1-x}Zn_x)_3O_{6.92(5)}$ (Y123) (3x=0.02, 0.05 and 0.10; T_c=81, 69 and 46 K), which were previously synthesized by annealing in reduced oxygen atmosphere in Ref. 10.

III. ZN-DOPING DEPENDENCE OF THE WIPEOUT NUMBER IN THE SUPERCONDUCTING STATE

No appreciable effect of nuclear spin diffusion in space nor in frequency is observed for Y124 and Y123, via (1) the stimulated spin-echo decay study [11], (2) pulse-strength H_1 dependence of the relaxation times, and (3) isotope dependence of the recovery curves. From these facts and the practically better fitting results [4], one can safely apply the magnetic impurity-induced NQR relaxation theory for Y124 and Y123.

Fig. 1 shows the Zn-doping dependence of the experimental recovery curves $p(t) \equiv 1 - M(t)/M(\infty)$ of the planar ^{63}Cu(2) nuclear magnetization $M(t)$ at 4.2 K. The solid curves are the least-squares fits of nonexponential function of,

$$p(t) = p(0)exp[-(3t/T_1)_{HOST} - N_c[e^{-3t/t_c} - 1 + \sqrt{3\pi t/t_c}\,erf(\sqrt{3t/t_c})]]. \quad (1)$$

The notations conform to the previous ones in Refs. 4 and 6. The fit parameters are $p(0)$, $(T_1)_{HOST}$, N_c and t_c. $(T_1)_{HOST}$ is a Cu nuclear spin-lattice relaxation time due to the host Cu electron spin fluctuation, N_c ($0 \leq N_c \leq 1$) is the wipeout number, t_c is an impurity-induced Cu nuclear spin-lattice relaxation time at an exclusion radius r=r_c centered around a Zn ion, and erf is the error function [7,8].

Eq. (1) is based on a *minimal model* for dilute alloys, which possesses at least two characteristic time constants $(T_1)_{HOST}$ and t_c, i.e., the host Cu electron spin correlation via a hyperfine coupling and the guest Zn-induced spin correlation via a longitudinal direct dipole coupling (the indirect couplings have also possible contributions), respectively. The wipeout effect in Eq. (1) is based on an "*all-or-nothing*" model. The exclusion radius r_c around each Zn is defined such that the nuclei inside the region of radius r_c centered around the Zn are unobservable whereas the nuclei outside the region are observable. This model seems to be highly simplified, but it is qualitatively consistent with the Zn-induced local moment model proposed by ^{89}Y NMR study above T_c [12].

Fig. 2(a) shows the Zn-doping dependence of the estimated N_c for Y124 and Y123 at T=4.2 K. Here, we assume no Zn(1), i.e., the in-plane Zn concentration x_{plane}=$2x$ for Y124 and x_{plane}=$3x/2$ for Y123. The magnitude of the wipeout number N_c for underdoped Y124 is larger than that for optimally doped Y123. The wipeout number N_c increases with Zn doping both for Y124 and Y123. However, the degree of the increase in N_c is slower than that expected from disappearance of the Cu nuclei inside a x_{plane}-independent neighboring shell by Zn. When I_{jnn} is the probability of finding a Zn ion at the j-th (j=1, 2, 3, 4) nearest-neighboring (nn) shell by Cu(2), then $I_{1nn} = I_{2nn} = I_{3nn}$=$4x_{plane}(1-x_{plane})^3$ and $I_{4nn} = 8x_{plane}(1-x_{plane})^7$. If the Cu(2) nuclei up to the j-th nn shells by Zn are unobservable, then $N_c^{1nn} = I_{1nn}(x_{plane})$ (dotted curve), $N_c^{2nn} = N_c^{1nn} + I_{2nn}(x_{plane})$ (short dashed curve), $N_c^{3nn} = N_c^{2nn} + I_{3nn}(x_{plane})$ (moderately long dashed curve), and $N_c^{4nn} = N_c^{3nn} + I_{4nn}(x_{plane})$ (long dashed curve). The increase of N_c for Y124 and Y123 is slower than that of $N_c^{4nn}(x_{plane})$ with increasing x_{plane}.

We approximate N_c by $x_{plane}\pi(r_c/a)^2$, a circle with an effective wipeout radius r_c. In Fig. 2(b), the estimated wipeout radius r_c/a is shown as a function of the in-plane Zn x_{plane} for Y124 and Y123. The wipeout region around each Zn shrinks with Zn doping both for Y124 and Y123. Fig. 3 shows the schematic illustration of the shrinkage of the wipeout region in the CuO_2 plane. The shaded area in the circle indicates the wipeout region around a Zn ion.

IV. T DEPENDENCE OF THE WIPEOUT NUMBER

The estimated values of $(1/TT_1)_{HOST}$, $1/\tau_1 T$ ($1/\tau_1 = \pi N_c^2/t_c$ [4, 6-8]), and N_c as functions of T for Y124 are shown in Figs. 4(a), (b) and (c), respectively.

(a)A small decrease of the normal-state $(1/T_1T)_{HOST}$ with Zn doping is consistent with the previous result form the analysis with N_c=0 [3,5,6]. This can be understood by suppression of the host antiferromagnetic correlation as a result from dilution effect of Zn on the magnetic CuO_2 network, similarly to the Zn-doped parent insulator $La_2Cu_{1-x}Zn_xO_4$ [13].

(b)A small decrease of the Zn-induced relaxation rate $1/\tau_1 T$ just below T_c is observed, which is in contrast to the increase of ^{7}Li Knight shift in the Li^+-doped Y123 [14]. From a relation of $1/\tau_1 T \propto K\tau$ (K is the Knight shift at the impurity site, and τ is the impurity magnetic correlation time), the life time τ of the impurity magnetic correlation decreases just below T_c.

(c)The value of N_c for x=0.01 above T_c is close to unity, so that it seems to be an overestimation. This is a shortcoming of the present analysis using Eq. (1), partially because the limitation due to mean impurity spacing is not taken into account. However, one can safely conclude that N_c for x=0.005 in Fig. 4(c) changes smoothly around T_c but does not diverge at T_c. This is sharply in contrast to the superconducting coherence length ξ_{SC}, which must diverge at T_c because the superconducting transition is the second order phase transition.

Fig. 5 shows the T dependence of the effective wipeout radius r_c/a for the Zn-doped Y124 (x=0.005, x_{plane}=0.01) estimated from a relation of N_c=$x_{plane}\pi(r_c/a)^2$. For comparison, various characteristic lengths in units of an in-plane lattice spacing a are plotted as functions of T; the experimental antiferromagnetic correlation length ξ_{AF} estimated from the ^{63}Cu(2) nuclear spin-spin relaxation rate (Gaussian decay rate) $1/T_{2G}$ [15], the superconducting coherence length ξ_{SC}=$\xi_{SC}(0)(1\text{-}T/T_c)^{-1/2}$ ($\xi_{SC}(T\text{=}0)$=$4.9a$ [16] and T_c=68 K) in the Ginzburg-Landau (GL) mean-field theory below T_c, the superconducting pair correlation length ξ_{dSC} in the self-consistent renormalization theory for a two-dimensional $d_{x^2-y^2}$-wave superconducting fluctuation model above T_c [17], and the above T_c superconducting coherence length ξ_{TDGL}=v_F/T (the effective Fermi velocity v_F=$1000a$) in a time-dependent GL theory (the dashed curve) [18]. The upward triangle indicates ξ_{SC}=$4.9a$ at T=0 extrapolated from an in-plane upper critical field H_{c2} just below T_c [16]. The downward triangles indicate an antiferromagnetic correlation length ξ_{in-gap} at T=10.5 K of Zn-induced in-gap spin fluctuations for the Zn-doped Y123 (x=0.02) [19]. Obviously, the magnitude and the T dependence of r_c for Y124 (x=0.005) are similar to ξ_{in-gap} and ξ_{AF} but not to ξ_{SC}.

Thus, one can associate the wipeout region with a locally enhanced antiferrmagnetic correlation region around Zn. The wipeout region may correspond to some virtual or real bound state induced by Zn in the antiferromagnetic background, so that it reflects the change of the host antiferromagnetic correlation length [20].

V. CONCLUSION

From the analysis of nonexponential planar Cu nuclear spin-lattice recovery curves, we observed an increase of the wipeout number N_c in the CuO_2 plane of Zn-doped Y124 and Y123 with Zn doping at 4.2 K. This is similar to the increase of the normal-state wipeout number N_c for the Zn-doped Y123 [9]. However, if N_c is approximated by $x_{plane}\pi(r_c/a)^2$, we found that the effective wipeout radius r_c around Zn shrinks with Zn doping. We associated the shrinkage and the T dependence of the wipeout region around each Zn with change in the host Cu antiferromagnetic correlation.

ACKNOWLEDGMENTS

This work was supported by New Energy and Industrial Technology Development Organization (NEDO) as Collaborative Research and Development of Fundamental Technologies for Superconductivity Applications.

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FIG. 1. The nonexponential recovery curves $p(t) \equiv 1 - M(t)/M(\infty)$ of the planar ^{63}Cu(2) nuclear magnetization $M(t)$ for Zn-doped Y124 and Y123 at T=4.2 K. The solid curves are fitted results based on Eq. (1).

FIG. 2. The wipeout number N_c versus the in-plane Zn content x_{plane} (a), and the effective wipeout radius r_c versus x_{plane} (b). We assume that Zn predominately substitutes for the planar Cu(2) site, that is, x_{plane}=$2x$ for Y124 and x_{plane}=$3x/2$ for Y123. See the text for the long (4nn), moderately long (3nn), short dashed (2nn), and dotted (1nn) curves in (a).

FIG. 3. The schematic illustration of the electronic state of the CuO_2 plane with Zn. The "arrow" represents the enhanced magnetic correlation in the wipeout region around a Zn ion (shaded circles).

FIG. 4. The estimated $(1/TT_1)_{HOST}$ (a), $1/\tau_1 T$ $(1/\tau_1 = \pi N_c^2/t_c)$ (b), and N_c (c) as functions of T for the Zn-doped Y124 (x=0.005 and 0.01).

FIG. 5. The T dependence of the effective wipeout radius r_c for the Zn-doped Y124 (x=0.005). ξ_{AF}, ξ_{SC} and ξ_{dSC} in units of an in-plane lattice spacing a are the antiferromagnetic correlation length [15], the superconducting coherence length below T_c, and the two-dimensional $d_{x^2-y^2}$-wave superconducting pair corrleation length above T_c [17], respectively. The dashed curve is the above T_c superconducting coherence length ξ_{TDGL} [18]. The upward triangle indicates an extrapolated ξ_{SC}=4.9a at T=0 [16]. The downward triagnles indicate the Zn-induced antiferromagnetic correlation length ξ_{in-gap} at T=10.5 K of Zn-induced in-gap spin fluctuations for the Zn-doped Y123 (x=0.02) [19].